

"Processing of Tar Sand Bitumens. Part I--
Thermal Cracking of Utah and Athabasca Tar
Sand Bitumens"

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Introduction

The tar sands deposits of Utah contain significant reserves of fossil energy. The bitumen, which is impregnated in sandstone, is a heavy, viscous, residue-like material and is not amenable to recovery by conventional primary or secondary techniques. Research and development efforts are currently being conducted in U.S. Government and in University laboratories to study the various recovery processes by hot water, fluid coking, and in situ combustion, etc. Additionally, efforts are being made to determine the optimum conditions for conversion of recovered bitumen to high quality products for use as fuel or the manufacture of chemicals.

The study of Utah tar sand bitumens has received relatively minor attention compared with Athabasca, Canada, bitumen. Virtually no literature exists regarding the processing of Utah bitumen whereas the Athabasca bitumen is being commercially processed. The thermal cracking of Asphalt Ridge and Sunnyside, Utah, bitumen was studied by Wenger, et al. (1) in the early 1950's. These studies were performed on bitumen which had been recovered via hot caustic water extraction followed by a solvent clarification step. This work provided valuable qualitative information about the yields and the nature of the products derived from Utah bitumens but results of these early studies are difficult to interpret in light of more recent information.

The thermal processing of Athabasca bitumen on the other hand, is well known and references to these studies are contained in an excellent overview by Camp. (2). The Great Canadian Oil Sands, Ltd., venture has selected delayed coking as the initial upgrading process and technology and processing conditions for the Athabasca bitumen are well established. However, Utah tar sands have been recently shown (3,4) to differ significantly from the Athabasca bitumen. This precludes the direct adaptation of existing technology and requires an additional research effort to identify the optimum processing conditions for this U.S. resource.

The work reported here represents our preliminary efforts to determine the processing characteristics and the value of the products derived from Utah tar sand bitumens. Thermal cracking was selected for our initial study because some variation of this general process will most likely be employed to upgrade bitumen to raw crude oil which can, in turn, be subjected to various hydrogenating/cracking sequences.

Our primary aim in this study was to provide a detailed comparison of the thermal cracking behavior of one bitumen to another and to compare tar sand bitumen generally with a petroleum residue. A direct comparison, under uniform, reproducible reaction conditions, provides a basis for determining the relative value of products derived as a function of the bitumen properties. In addition, the results of this study will provide a base from which to evaluate effects of varying the reaction conditions on yield and product properties.

In this study we subjected three Utah bitumens, one Athabasca bitumen and a Wilmington, Calif., petroleum residue to batch-type destructive distillation at atmospheric pressure and under an inert atmosphere. Properties and composition of the native bitumen are summarized and results of bench-scale thermal cracking are discussed in terms of the bitumen composition. Properties of the products from Utah bitumen are compared with that from Athabasca and the petroleum residue. General conclusions about the relative value of Utah bitumens as feeds for thermal cracking are made.

Experimental Procedure

Description of Samples - Four tar sand bitumens and one petroleum residue were thermally cracked in this work. Two tar sand bitumens, the Athabasca, Canada and P.R. Spring, Utah, were identical to the samples used in previous studies of one of the authors of the compound type analysis. (3,4) The Asphalt Ridge and Tar Sand Triangle, Utah, samples were similar to those two used in this previous work. Because property and elemental analysis data corresponded closely between the previous samples and present ones, we assumed bitumen composition to be respectively identical for purposes of interpreting the cracking yield data. The tar sand bitumens were extracted from the tar sand with benzene, filtered through 4.0 to 5.5 μ fitted glass filters, and flash distilled until final conditions of 75° to 80° C and 4 to 5 torr had been maintained for 1 hour.

The petroleum residue was a 485° C distillation residue from a Wilmington, Calif., crude oil. This sample was identical to that reported on earlier (3) and in conjunction with analysis resulting from American Petroleum Institute-U.S. Bureau of Mines Research Project 60. (5) This sample was determined to be the most similar to the P.R. Spring bitumen in terms of physical properties and composition and was included to establish a point of reference for our present work.

Description of apparatus- The Vycor thermal cracking reactor consisted of a common distillation flask design with the sidearm turned downward at the end. Heat was applied to the pot by use of a beaded heater and to the head and sidearm by use of heating tape. Temperature was monitored inside and outside of both the pot and head with thermocouples. The top of the reactor was closed off with a 19/38 joint which had been fitted with thermowells. Liquid products were collected in cold-finger type condensers using dry-ice/methanol for the final coldfinger coolant.

Description of procedure - Approximately a 10 g-sample of bitumen was pyrolyzed at a heating rate of about 5° C/min to an end point of 625° C. Reactor and condensers were preweighed and coke and liquid product yields were determined directly by weight. Gas volume rate production was determined by measuring the volume of water displaced

at atmospheric pressure and subsequently corrected to STP conditions. Weight of gas produced was estimated by difference and by analysis of gases produced.

Analysis of bitumen and products - Gas analysis was accomplished by GC using a 1/16-inch by 16-foot activated alumina column and by mass spectrometry. Elemental analysis and physical properties of the bitumen and products were determined by conventional means by U.S. Government and commercial laboratories. Heating value (H) was calculated from the formula (6)

$$H(\text{Kcal/K}_g) = 8,400(C) + 27,765(H) + 1,500(N) + 2,500(S) - 2,650(O).$$

Simulated distillation of bitumen and liquid products was accomplished by G.L.C. by procedures fully described elsewhere (7), using a 1/4-inch by 18-inch column packed with 5 percent UCW-98 on 60 to 80-mesh silanized chromosorb W. TGA and DTA were performed on a Mettler thermogravimetric analyzer. New infrared data reported here were gathered using a Beckman model 4860.

Results and Discussion

Bitumen, Composition and Properties

The extracted tar sand bitumens were characterized by methods adapted from those developed for the study of high boiling petroleum distillates and residues. Results of this study, which was conducted at the Laramie Energy Research Center (ERDA), will soon be published, (4) and are summarized here to provide an understanding of the nature of the bitumen. The elemental analysis and selected physical properties are given in table I. Comparison is made of the two sets of bitumens for which the sample used for cracking was similar to, but not identical to, that characterized. In the case of the Tar Sand Triangle sample, the two samples are nearly identical whereas some differences are noted for the Asphalt Ridge samples.

Results of our previous studies (4) have shown marked similarities between the two Uinta Basin samples (Asphalt Ridge and P. R. Spring) on the one hand and the Athabasca and Tar Sand Triangle on the other. These similarities extend beyond the elemental analysis and physical properties and include compound type analysis and boiling point distribution as well.

Comparison of the property and distillation data for the Uinta Basin samples with the high sulfur bitumens reveals a higher hydrogen and nitrogen content, lower sulfur, higher molecular weight and viscosity, and lower content of volatile material. These results are interpreted in conjunction with the compound type analysis to mean that Uinta Basin samples have a higher percentage of high boiling and residual saturated hydrocarbons with the aromatic portion reserved appreciably for the high boiling residue fraction. Conversely, the Tar Sand Triangle and Athabasca bitumens are comprised of generally lower molecular weight material with the aromatics and heteroatoms making up a comparatively large percentage of the volatile material.

There are a few notable compositional differences between the Wilmington residue and the tar sand bitumens which are likely to

Table I - Elemental analysis and physical properties
of bitumens*

Property	ATH ⁽⁴⁾	TST ₁ ⁽⁴⁾	TST ₂	AR ₁ ⁽⁴⁾	AR ₂	PRS ⁽⁴⁾	WIL ⁽⁷⁾
Carbon (wt-pct)	82.6	84.0	84.2	85.3	85.5	84.4	85.4
Hydrogen	10.3	10.1	10.2	11.7	11.3	11.0	10.6
Nitrogen	.47	.46	.52	1.02	1.03	1.0	1.08
Sulfur	4.86	4.38	4.48	.14	.42	.75	2.08
Oxygen	1.8	1.1	1.7	1.1	1.6	3.3	.8
C/H atomic ratio	.674	.695	.693	.611	.634	.641	.674
Average molecular wt. (VPO-benzene)	568	578	542	668	666	820	640
API gravity	11.6	11.1		14.4	12.0	10.3	8.6
Carbon residue (Ramsbottom), wt-pct	16.1	21.6	17.0	3.5	9.1	12.5	18.0
Asphaltene (pentane) wt-pct	16.4	26.0		3.4		16.0	12.8
Viscosity (poise) 77° F (cone-plate at 0.05 sec ⁻¹)	6,380	12,900	29,500			325,000	99,500
Pct volatile material (535° C)	60.4	54.5		50.3		49.9	(est)25

* ATH - Athabasca
TST - Tar Sand Triangle (previous study)
TST₁ - Tar Sand Triangle (this study)
TST₂ - Tar Sand Triangle (this study)
AR₁ - Asphalt Ridge (previous study)
AR₂ - Asphalt Ridge (this study)
PRS - P. R. Spring
WIL - Wilmington

influence the chemical behavior but are not apparent in the gross properties. Although the total heteroatom content of the Uinta Basin bitumen is similar to that of the Wilmington residue, there is a greater tendency (as a result of geochemical forces) to concentrate the heteroatoms in the high molecular weight compounds of the bitumens. Additionally, there is considerably more material associated with a given quantity of heteroatoms for comparable initial boiling point residues indicating that the average molecular weight for a 530° C residue of the Uinta Basin samples may be 1-1/2 to 2 times that of a 530° C Wilmington sample. The Athabasca and Tar Sand Triangle samples exhibit lesser tendencies to concentrate heteroatoms in the residue portion than the Wilmington residue. The comparative upper molecular weight of these samples is not known.

Quantitative Results of Pyrolysis

Product yields from the batch-type thermal cracking of the five bitumens are given in table II. Gas yields, which range from 4.8 to 7.5 percent, do not exhibit any obvious correlation with bitumen properties. The majority of the gas was produced above 500° C when condensate production was tapering off. The percent gas yield and the average molecular weight of the gas ran roughly parallel to each other.

Table II - Product yields

Product	ATH	TST	AR	PRS	WIL
Gases (C ₅ and lighter)	7.52	5.31	4.80	7.41	6.03
Liquid condensate (C ₆ -535° C)	76.52	72.82	82.85	76.05	77.04
Coke	15.96	21.87	12.35	16.54	16.93

Liquid condensate yields exhibit a good inverse correlation with carbon hydrogen atomic ratio. A secondary correlation appears to exist with molecular weight; for a given C/H ratio, the higher the molecular weight, the lower the yield. The Asphalt Ridge sample, which is a likely candidate for early commercial development, gave a high yield of almost 83 percent condensate.

Coke yield, which is directly related to carbon residue determination, also correlates well with the asphaltene content. Asphaltenes derived from tar sands are probably similar to petroleum asphaltenes in that they represent the high molecular weight, highly aromatic molecules present. These molecules contain significant quantities of heterocyclic structures with generally low alkyl and naphthenic carbon contents. These structural features explain their resistance to pyrolysis and their ability to form coke. Although the correlation between coke yield and asphaltene content is strong, a one-to-one precursor-product mechanism is not suggested. Many nonasphaltene aromatic molecules probably undergo polymerization and condensation as well, and asphaltenes can rupture to form lighter products. Comparison of the results of cracking a total bitumen with results from a deasphalted bitumen should prove interesting.

In order to provide assurances that yield and analytical results would be meaningful, it was imperative that high material and elemental

balance be achieved. This was one of the principal considerations in the experimental design. Table III shows the elemental balance for the Wilmington sample. Other bitumens showed similar results. The data illustrate that carbon and hydrogen recoveries were excellent, while nitrogen and sulfur were slightly low. Material balance was better than 99 percent in all cases.

Table III - Elemental balance (Wilmington residue)

	Wt-pct (O free basis)	
	Starting bitumen	Analysis of recovered products
Carbon	86.1	86.7
Hydrogen	10.7	10.8
Nitrogen	1.1	0.9
Sulfur	2.1	1.6

Characterization of Products

Gaseous products were analyzed by gas chromatography and mass spectrometry. All gaseous products showed the typical constitution of a pyrolysis gas. An example of mass spectral analysis for the products derived from the high sulfur Tar Sand Triangle bitumen is given in table IV. The oxygenated compounds are believed to be derived from bitumen oxygen compounds. Gases derived from the other bitumens were of a similar constitution with that from Asphalt Ridge having a relatively high concentration of olefins and that from the Wilmington having a relatively high concentration of methane.

Table IV - Composition of Tar Sand Triangle gases

Compound	Mole, pct (Helium-free basis)	Compound	Mole, pct (Helium-free basis)
Hydrogen	14.3	Cyclopentene	0.1
Methane	47.3	Pentenenes (noncyclic)	.7
Ethylene	1.6	Isopentane	.3
Ethane	10.9	n-pentane	1.3
Propylene	3.1	Ammonia	.7
Propane	5.5	Hydrogen sulfide	5.0
1,3-butadiene	0.1	Carbon monoxide	3.9
Butenes	2.6	Carbon dioxide	.4
Iso-butane	0.0		
n-butane	2.2	Total	100.0

Liquid products were characterized by elemental analysis, physical properties and application of the n-d-M method (9) for carbon type and ring structure, infrared spectroscopy, and simulated distillation. Elemental analysis and physical properties are given in table V. Generally, the bulk properties are remarkably similar for all samples. Carbon/hydrogen ratios exhibit moderate variation with values grossly following that in the original bitumen. The Tar Sand Triangle condensate exhibited a substantial increase in hydrogen content over the native bitumen while other samples exhibited less marked enrichment.

Table V - Liquid condensate properties

	ATH	Bitumen			
		TST	AR	PRS	WIL
Carbon (wt-pct)	84.7	85.2	87.1	86.5	86.5
Hydrogen	11.3	11.6	12.0	12.1	11.7
Nitrogen	.19	.16	.58	.57	.43
Sulfur	3.75	2.68	.32	.29	1.43
Oxygen	0 - trace	0 - trace	0 - trace	0 - trace	0 - trace
C/H atomic ratio	.631	.616	.615	.598	.618
Average molecular wt (VPO benzene)	279	280	282	280	313
Specific gravity (20/20)	.923	.910	.898	.895	.920
API gravity	21.9	24.0	25.8	26.5	22.3
Refractive index -n ²⁰	1.5191	1.5130	1.5106	1.5053	1.5174
Heating value (Btu/lb)	18,632	18,803	19,084	19,153	19,002

Sulfur and nitrogen contents reflect the relative concentrations of these elements present in the native bitumen. Oxygen has been completely removed, presumably as water and the oxides of carbon. Small amounts of water were produced at reaction temperatures of 350° to 400° C. Molecular weights are nearly identical for the tar sand bitumens while API gravity and refractive index show minor variations. The significance of the nearly constant physical properties can be seen in the results of the n-d-M analysis.

Application of the Van Ness and Van Weston (9) n-d-M method for estimating carbon structure was made with the recognition that these correlation charts were developed for virgin olefin-free petroleum samples. The presence of either cyclic or straight-chain olefins in a sample would probably add semiquantitatively to the aromatic carbon content largely at the expense of the naphthenic carbon. The percent of olefin carbon present in condensate products derived from the destructive distillation of Asphalt Ridge bitumen (1) and delayed coking of Athabasca bitumen (10) range from 10 to 12 percent. The n-d-M carbon type analysis was adjusted for the olefin content by subtracting the olefin carbon from the aromatic carbon and assuming that all olefins were present in a ring system. Typical ranges of carbon type and ring type are given in table VI. Inspection of the detailed data for products from each bitumen revealed minor differences, with the naphthenic carbon exhibiting the highest variability. These analyses further confirm the structural similarity of the pyrolysis condensates that were indicated by the similarities in the properties.

Table VI - Carbon and ring type analysis of condensates

Type	Atomic pct of carbon
Aromatic carbon	18-20
Naphthenic carbon (saturated)	9-16
Olefin carbon	10-12
Paraffinic carbon	55-60
Aromatic rings/molecule	0.7
Naphthenic-olefin rings/ molecule	0.8-1.2

Infrared spectra was employed to obtain additional qualitative information about the liquid condensate products. Virtually no adsorption exists in the free or bonded O-H or N-H region. The C-H region showed virtually no aromatic C-H and the intensity of the methyl C-H nearly equaled that of the methylene C-H. A broad, ill-defined adsorption peak centering about 1,705 cm^{-1} indicative of the presence of carbonyl compounds, is probably due to ketone artifacts generated upon exposure of the sample to air. A fairly sharp band centering about 1,640 cm^{-1} was attributed to unconjugated olefins. The existence of asymmetry on the high frequency side of the aromatic band (1,603 cm^{-1}) was attributed to the presence of conjugated olefins. Assignments were not made for several strong bonds present from 1,000-700 cm^{-1} .

The boiling point distribution was determined from simulated distillation by gas-liquid chromatography. Results are given in table VII. In this analysis significant differences are obvious. This is rather surprising in light of the similarities in the other properties

Apparently an "average structure" is defined by thermodynamic considerations but the actual distribution of molecular sizes relates back to some yet unknown structural feature of the feed bitumen. The Wilmington condensate is noticeably heavy which is consistent with its higher molecular weight and slightly higher degree of ring condensation.

Table VII - Simulated distillation yields, wt-pct

	ATH	TST	AR	PRS	WIL
Gasoline C ₅ ⁺ - 200° C	7.5	7.2	11.9	10.4	8.6
Kerosine 200-275° C	12.9	11.5	19.9	14.7	11.6
Gas oil 275-325° C	13.7	13.0	16.9	12.8	8.1
Heavy gas oil 325-450° C	48.0	51.4	34.0	46.6	38.5
Vacuum gas oil 450-535° C	17.9	16.9	17.3	15.6	28.2
Subtotal	100.0	100.0	100.0	100.0	95.0
Residue	0-4%	0-3%	0-5%	0-2%	5-10%

The coke was characterized by elemental analysis and heating value (table VIII). All of the cokes had a shiny appearance with infrequent pores. Coke from the Uinta Basin samples are extremely low in sulfur. Incomplete combustion in analysis is thought to cause low carbon values in some samples. Heating values were calculated from actual percentages because oxygen content is not known.

Table VIII - Coke analysis

	ATH	TST	AR	PRS	WIL
Carbon (wt-pct)	88.6	87.7	87.9	87.7	89.8
Hydrogen	2.5	2.8	3.0	2.6	2.9
Nitrogen	1.8	1.5	2.9	2.9	3.0
Sulfur	6.0	6.2	0.4	0.5	1.5
Heating value (Btu/lb)	14,960	14,950	14,860	14,720	15,165

Discussion of Results

The experimental design optimized sample definition. This procedure provides a base for comparison of results which can be derived from visbreaking, delayed coking, catalytic cracking and catalytic hydrogenation experiments. Although reaction conditions do not simulate commercial coking operations preliminary interpretation can be made of the thermal cracking characteristics of Utah tar sand bitumens. By relating results from Utah samples to those derived for Athabasca and a representative petroleum residue comparison is established with commercially processed samples. Results for the Athabasca sample compare favorably with literature results (2) with gas productions and compositions being similar to delayed coking operations while liquid yields are higher (77 vs 70) and coke yields are lower (16 vs 22) in the present study. These results are explained by the longer residence time under higher hydrocarbon partial pressures which enhances the condensation reactions.

Certain observations regarding the rate of light gas and liquid condensate production deserve attention. Plots of the rate of gas production as a function of temperature revealed a trimodal curve with the major rate of gas production occurring about 490° C with less prominent peaks occurring at 555° C and 610° C. These data suggest that several distinct molecular types may be present with discrete decomposition temperatures. Analysis of gases produced as a function of temperature revealed the expected increase in olefin to saturated hydrocarbon ratio but exhibited no clear enhancement of one component over another. At higher temperature the C_2-C_5/C_1 ratio was enhanced. Generally, more than half the gas production occurred after liquid condensate production had ceased (ca. 525° C). The peak gas production occurred some 40° C after peak condensate production. This suggests that precursors to light gases are largely short alkyl (or naphthene) groups attached to larger nonvolatile molecules. Cracking of medium volatility molecules or long chain alkyl substituents to light gases would give rise to simultaneous condensate production which is not observed.

Additional insight into the cracking behavior of these bitumens is obtained from thermal gravimetric analysis (TGA). Typically, the differential TGA curve (change in rate of weight loss vs temperature) exhibits a broad peak at 275° to 350° C followed by a well defined peak at 435° to 460° C. Comparison of TGA results with those of the simulated distillation, which determines volatility at short high-temperature residence times, reveal that initial thermal cracking may be taking place at temperatures as low as 150° C with appreciable cracking existing at 300° C. The susceptibility to thermal cracking was particularly apparent with the Wilmington sample where over 45 percent TGA weight loss had been experienced by 375° C when the nominal IBP was 485° C. Conversely, visual observation of the production of liquids from tar sand bitumens in the bench scale experiments showed substantially less liquid production at 375° C than was expected based on the simulated distillation data. The implication in these results is that even though thermal cracking apparently occurs at lower temperatures, mass transfer effects may be a limiting factor.

The thermal reactivity of tar sand bitumens is probably ultimately related to the overall severity of the geochemical maturation process. In this regard the Wilmington sample appeared noticeably more reactive than did the tar sand bitumens. This leads to the speculation that maturation conditions have been more severe for tar sands compared to the Wilmington crude oil. This factor will probably affect the optimum reaction conditions for upgrading bitumen to synthetic crude oil.

The work reported here is the result of a preliminary investigation at one set of reaction conditions. For this reason, further discussion of the mechanistic implications of our analytical work is reserved for future publications when the effects of varying the reaction conditions can also be discussed. Suffice it to say that knowledge of the total aromatic, naphthenic, paraffinic content and molecular weight data will not be adequate to explain the results. More subtle factors which may have significant effects on yield and yield structure include degree and type of alkyl substitution, the type of ring condensation and the presence of coke promoting heteroatoms such as pyridine type nitrogen. The model compound pyrolysis study of Madsen and Roberts (11) relates to the effects of these compositional features.

Conclusions

The raw liquid condensate from the low sulfur Uinta Basin samples show good potential for hydrotreating to produce a high quality synthetic crude oil amenable to conventional processing. Compared to the Athabasca bitumen sample, the Uinta Basin products are low in sulfur, high in hydrogen, are of comparable olefin content and are produced in higher yields. These comparative features will significantly enhance the value of this tar sand resource. Generally, the high sulfur bitumen from Utah, i.e., Tar Sand Triangle, will produce a similar product in thermal processing to the Athabasca bitumen.

Our preliminary evaluation of the thermal cracking of Utah and Athabasca tar sand bitumens showed that (1) conversion is primarily a function of hydrogen content, (2) bitumens show appreciable cracking at 300° C with maximum cracking at 450° C, (3) condensable product character, although considerably uniform from one bitumen feed to another, tends to correlate with bitumen character, (4) products derived from tar sand bitumens compare favorably with those derived from a representative petroleum residue, (5) coke generated from Uinta Basin, Utah, samples is extremely low in sulfur; (6) hydrogen demands in hydrotreating processes will probably be lower for Utah samples than for Athabasca samples.

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